

## REMARKS/ARGUMENTS

The Office Action in the present case was mailed on October 28, 2004, making a response due on or before January 28, 2005. Since this response is being timely submitted, no further fee is thought to be due at this time. If any additional fee is due for the continued prosecution of this application, please charge the same to Applicant's Deposit Account No. 50-2555 (Whitaker, Chalk, Swindle & Sawyer, LLP).

The present case is a divisional of prior serial number 09/911,021, now issued as U.S. Patent No. 6,790,363. In that case, a restriction requirement was issued forcing an election between Claims 1-6 drawn to an adsorbent material; Claims 7-13 drawn to a method for making a coated zeolite; Claims 14-19 and 24-32, drawn to a method for removing arsenic from water; and Claims 20-23 and 33-37 drawn to a filtration unit. In the parent case, Applicant elected to prosecute Claims 14-19 and 24-32 which were drawn to the method of removing arsenic from water. This divisional application was filed with a Preliminary Amendment which canceled all but claims 7-13 drawn to the method for making a coated zeolite which claims are currently being examined.

The Examiner has substantively rejected Applicant's Claims 7-13 under 35 U.S.C. §102(b) as being anticipated by Vergani et al. (U.S. Patent No. 5,716,588). Reconsideration of the rejected claims is respectfully requested in view of the arguments which follow.

The Examiner admits that Vergani fails to disclose that the Mn-Fe is "nanophase" as required by Applicant's claims. However, the Examiner argues that since the reference discloses the use of the same starting materials and prepares the product using the same process, that the same nanophase product would inherently be produced (Office Action of October 28, 2004, paragraph 2). In other words, the Examiner argues that the "nanophase" characteristic of the product being claimed by Applicant is an inherent characteristic of the prior art Vergani product. However, for the reasons which follow, Applicant respectfully traverses this conclusion of the Examiner. In fact, the Vergani et al. reference fails to anticipate Applicant's invention, as defined in Applicant's remaining claims.

In order to differentiate the process of Vergani et al. and the product which necessarily results therefrom, the Applicant will first summarize the Vergani et al. process:

### The Vergani Process

1. 96 g of  $\text{FeCl}_3$  and 41.7 g of  $\text{MnCl}_2$  are dissolved in 0.5 L of distilled water and the solution is heated to 50°C to dissolve:
  - a.  $\text{FeCl}_3$ , 6 $\text{H}_2\text{O}$  MW = 270 and  $\text{MnCl}_2$  4 $\text{H}_2\text{O}$  MW = 196.
  - b. Mn/(Mn+Fe) molar ratio is 0.35.
2. 7.5 g of pulverized zeolite 13X is added and the solution is diluted to 1L and heated at 60°C for 5 min.
3. Solution cooled to 40°C.
4.  $\text{NH}_4\text{OH}$  is added to increase the pH to 7.5 to form precipitates.
5. The suspension heated to 60°C for 5 minutes and cooled to 30°C.
6. For improved yield concentration  $\text{NH}_4\text{OH}$  was added to bring the pH of the precipitate to 9.
7. For 5 minutes, the solution was centrifuged and collected precipitates repeatedly washed with aqueous  $\text{NH}_4\text{OH}$  having a pH of about 8.
8. 4 g of amorphous Si was added to the precipitate to improve the mechanical consistency.
9. After drying at 75°C for 24 hrs the mixture was calcined at 700°C for about 4 hrs to convert Fe and Mn hydroxides in to their respective oxides.
10. The Fe and Mn getter materials were further treated at 400°C for 4 hrs using steady stream  $\text{H}_2$  gas, additional 400°C for 4hr with steady stream Ar gas. The getter material was cooled in Argon stream.

### Differences in Vergani et al and Applicant's Process and Product:

Two objectives are essential to Applicant's process:

- 1) Producing A Nanophase Material: This is due to the fact that higher surface areas ( $>450\text{ m}^2\text{ g}^{-1}$ ) will result in increased As(V) sorption. Heating the oxide ( $>100^\circ\text{C}$ ) at any stage of the process will result in crystalline Mn-Fe oxide phase, thereby considerably decreasing the As(V) sorption.
- 2) Substitution of Mn in the Fe oxide structure without forming any discrete Mn oxide phase. The Mn in the structure will oxidize As(III) to As(V) and the nanophase Fe oxide will sorb As(V).

In Applicant's Specification, nanophase Mn-subsituted Fe oxide coated zeolites were referred to as:

- Mn ferrihydrite (Specification, page 3, line 22).
- Natural zeolite coated with nanophase Mn-Fe oxide, NZNPF, (Specification, page 4, line8).
- Mn-subsituted nanophase Fe oxide (Specification, page 4, lines 1-2).

These exact terms were intended to describe a unique product which is distinguishable in its characteristics from prior art coated zeolites as well as the “getter materials” of Vergani et al. These particular products of Applicant’s process differ from the products which will necessarily result from the Vergani process as summarized above. For example, a 1% Fe oxide coating was sought by Applicant as all the nanophase Mn-Fe is coated on to the zeolite surface with little to no uncoated precipitated Mn-subsituted nanophase Fe oxide. See, page 7, lines 6-7 of Applicant’s Specification: “More specifically, NZNPF comprises 1% Fe oxide.” This is because the cation exchange capacity of natural zeolite is in the range of 100 to 250 milliequivalent per 100 g, depending on the particle size, which translates to Fe(III) maximum sorption in the range of 1.8% to 4.6% on the zeolite surfaces. In the process stated in this patent, the zeolite particles in the range of 0.4 mm to 0.8 mm will have up to 2% Fe and particles in the range of 0.1 mm to 0.2 mm will have up to 4% Fe in the form of nanophase oxide coated on their surfaces. There will be negligible to no excess free or pure oxides (without zeolite coating). This is extremely critical for Applicant’s intended use of the

products produced by the claimed process as As removal agents. The presence of free oxide will clog the water flow in the column. Also, the presence of excess of MnO<sub>2</sub> will clog the column resulting in decreased/water flow, and the absorption of As anions (H<sub>2</sub>AsO<sub>4</sub><sup>-</sup> and HAsO<sub>4</sub><sup>2-</sup>) will be greatly reduced because MnO<sub>2</sub> has negative surface charges.

To the best of Applicant's knowledge, the intent of Vergani et al., was to coat the zeolite with elemental Fe and Mn to form getter material to remove O<sub>2</sub> from ammonia. The Vergani patent does not, therefore, teach the synthesis of nanophase Mn-substituted Fe oxide, a discrete single phase. The term "Co-precipitation" in Col. 4, line 26, of Vergani refers to formation of two discrete phases. Similarly, continuing at Col. 4, lines 33 to 39, the reference teaches "iron and manganese precipitation"; the authors' ultimate goal is, "To make metallic elements dispersed on an inert support by sequentially heating under hydrogen and argon" which is contrary to Applicant's objective which is to make nanophase Mn-substituted Fe oxide coated on a zeolite support. In fact, the heating steps of Vergani would significantly decrease the surface area and arsenic sorption capacity of Applicant's product; additionally, it will render the oxide to form a crystalline Fe oxide phase (hematite). Thus, the Vergani et al. process does not anticipate Applicant's claimed process because first and foremost, the product produced is not a "nanophase" material as claimed by Applicant. Also, Applicant's final product is not a "getter material" as described by Vergani.

Applicant's claimed process is concerned with making Mn-substituted Fe oxide which is preferably coated on a natural zeolite. Particular requisites apply to Applicant's process which might be entirely irrelevant to the process and product of Vergani. For example, Applicant's preferred process uses a ratio of Mn to Fe in solution in which the Mn/(Mn+Fe) molar ratio is 0.1 or less. See, for example, page 12, lines 2-4 of Applicant's Specification: "Then a known amount of MnCl<sub>2</sub> was added to the Fe solution to get a Mn/(Mn + Fe) molar ratio of 0.1." In the case where the Mn/(Mn+Fe) ratios are >0.1, a separate Mn-phase Jacobsite and/or amorphous Mn oxide will precipitate under alkaline conditions. Our intent is for Mn(III) to replace Fe(III) in the nanophase Fe oxide structure to form a single phased mineral. The nanophase Mn substituted Fe oxide is amorphous containing high surface and arsenic adsorption capacity

The excess of Mn oxide prepared by Vergani et al., will significantly decrease the As(V) sorption and clog or impede the water flow because of its fine size particles. Our propose is to substitute the Mn in the structure to oxidize As(III) to As(V), and Fe oxide to capture the As(V). This will not reduce the adsorption capacity of the Mn-substituted nanophase Fe oxide

The above discussions is intended to emphasize the very different nature of the Vergani product from the product which is produced by Applicant's process. The brick red color of the coated zelite further confirms that Mn is substituting for nanophase Fe oxide structure. Otherwise, a green varnish color or brown colored coating will be formed.

Natural zeolite contains  $\text{CaCO}_3$  and heating to 60°C could precipitate the dissolved Fe and Mn to their respective carbonate minerals. This will in turn affect the final getter material. Therefore it is essential that in the Vergani et al. process the getter material is manufactured using zeolite 13X, a synthetic zeolite. Applicant's process, on the other hand, can use natural zeolites.

The present discussion is intended to emphasize the very different nature of the Vergani product from the product which is produced by Applicant's process. Applicant's process can be carried out at room temperature. In this regard, note Step 9 in the above discussion of the Vergani process: Drying the sample at 75°C for 24 hrs could revert the oxide to goethite ( $\text{FeOOH}$ ), a crystalline Fe oxide and likely to birnessite ( $\text{MnO}_2$ ), a crystalline Mn oxide. Finally, heating to 700°C will yield corresponding oxide. The goethite will form hematite and birnessite to nsutite, pyrolusite or todorokite, a stable  $\text{MnO}_2$  oxide (McKenzie, 1977, attached).

Finally, in the Vergani process, a stream of  $\text{H}_2$  at 400°C is passed through the material to form metallic Fe and Mn supported zeolite.

Applicant has amended remaining independent Claim 7 to specify "the ratio of Mn to Fe in solution as determined by the formula  $\text{Mn}/(\text{Mn} + \text{Fe})$  being 0.1 or less;" Note, in this regard, the  $\text{Mn}/(\text{Mn}+\text{Fe})$  molar ratios utilized in Vergani et al.'s process:

In Example 1, page 6 and lines 8 to 12. The Mn/(Mn+Fe) ratio is 0.35. In this system, a discrete Mn oxide phase will be formed in addition to Fe oxide phase. Also, the %Fe based on zeolite weight basis is 276, which results in excess of uncoated Fe oxide and Mn oxide.

In Example 2, page 6 and lines 63 to 64). The Mn/(Mn+Fe) ratio is 0.56. In this system, a separate Mn oxide phase will be formed in addition to Fe oxide phase. The %Fe based on zeolite weight basis is 56, which results in excess of uncoated Fe oxide and Mn oxide.

In Example 3 (page 7) the 7:1 calculated Fe to Mn ratio is converted to Mn/(Mn+Fe) molar ratio of 0.11, which will result in formation of a discrete Mn oxide phase. Further, heating to 560°C will result in crystalline discrete Fe and Mn oxide phases. The %Fe based on zeolite is 256, which results in excess of uncoated Fe oxide and Mn oxide.

In Example 4 (page 7) the 0.93:1 calculated Fe to Mn ratio is converted to Mn/(Mn+Fe) molar ratio of 0.46, which will result in formation of a discrete Mn oxide phase. Further, heating to 560°C will result in crystalline discrete Fe and Mn oxide phases. The %Fe based on zeolite is 146, which results in excess of uncoated Fe oxide and Mn oxide.

While Applicant could enter amendments related to additional differences in the starting materials and process steps outlined above, Applicant would submit that the Examiner should be willing to grant a claim commensurate in scope with the invention. Here, for the multitude of reasons presented above, the Vergani et al. reference fails to teach a method for producing a “nanophase” material as explicitly claimed by Applicant. This is evident from the fact that Vergani fails to utilize the molar ratio of starting materials now specified in Applicant’s amended claims. As a result, independent Claim 7 should be allowable over the Vergani reference. The remaining claims all depend from Claim 7 and should similarly be allowable.

Based upon the above arguments, Claims 7-13 are thought to be allowable over the art of record and an early notification of the same would be appreciated.

Please charge any additional fees which may be due for the continued prosecution of this application to Applicant's Deposit Account No. 50-2555 (Whitaker, Chalk, Swindle & Sawyer, LLP).

Respectfully submitted,

Date: Jan 27, 2005

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